IN THE UNITED STATES PATENT AND TRADEMARK OFFICE Before the Board of Patent Appeals and Interferences

> Atty Dkt. 461-62 M#

C# TC/A.U.: 1754

Examiner: E. Johnson

Date: November 22, 2004

KOIKE et al

Serial No. - 09/546,227

In re Patent Application of

Filed: April 10, 2000 Title:

A CERAMIC SUPPORT CAPABI

SUPPORTING A CATALYST...

Mail Stop Appeal Brief - Patents

Commissioner for Patents

P.O. Box 1450

Alexandria, VA 22313-1450

Sir:

☐ Correspondence Address Indication Form Attached.

NOTICE OF APPEAL

Applicant hereby appeals to the Board of Patent Appeals and Interferences \$ from the last decision of the Examiner twice/finally rejecting (\$340.00) applicant's claim(s).

An appeal **BRIEF** is attached in the pending appeal of the above-identified application (\$ 340.00)

340.00

Credit for fees paid in prior appeal without decision on merits

330.00)

A reply brief is attached in triplicate under Rule 41.41

(no fee)

0.00)

Petition is hereby made to extend the current due date so as to cover the filing date of this paper and attachment(s) (\$110.00/1 month; \$430.00/2 months; \$980.00/3 months; \$1530.00/4 months)

10.00 SUBTOTAL

Applicant claims "Small entity" status, enter ½ of subtotal and subtract

-\$(

☐ "Small entity" statement attached.

SUBTOTAL 10.00

month extension previously paid on Less

> **TOTAL FEE ENCLOSED \$** 10.00

-\$(

Any future submission requiring an extension of time is hereby stated to include a petition for such time extension. The Commissioner is hereby authorized to charge any deficiency, or credit any overpayment, in the fee(s) filed, or asserted to be filed, or which should have been filed herewith (or with any paper hereafter filed in this application by this firm) to our Account No. 14-1140. A duplicate copy of this sheet is attached.

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CC:lmr

NIXON & VANDERHYE P.C.

By Atty: Chris Comuntzis, Reg. No. 31,097

Signature:

901769

#### IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re Patent Application of

KOIKE et al

Serial No. 09/546,227

Filed: April 10, 2000

For: A CERAMIC SUPPORT CAPABLE OF

SUPPORTING A CATALYST...

Atty. Ref.: 461-62

Group: 1754

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Mail Stop Appeal Brief - Patents

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Sir:

#### **APPEAL BRIEF**

Appellants hereby appeals the Final Rejection of August 20, 2003.

Adjustment date: 11/23/2004 LWDNDIM1 03/18/2004 AWDNDAF1 00000048 09546227

**REAL PARTY IN INTEREST** 

The real parties in interest are Appellants, Nippon Soken, Inc. and Denso Corporation.

#### RELATED APPEALS AND INTERFERENCES

The Appellants and the undersigned are not aware of any related appeals or interferences which will directly affect or be directly affected by or have a bearing on the Board's decision in this appeal.

11/23/2004 LWDNDIH1 00000075 09546227

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#### STATUS OF THE CLAIMS

Claims 73-78, 80-99, 102 and 103 remain pending in this application. Claims 73-78, 80-86, 94-99 and 102 stand rejected by the Examiner, the rejections of which are appealed. Claims 87-93 and 103 have been allowed. All of the claims are presented in the Appendix I to this Brief.

### STATUS OF ANY AMENDMENT FILED SUBSEQUENT TO ANY FINAL REJECTION

No amendments have been filed subsequent to the Final Rejection.

#### SUMMARY OF CLAIMED SUBJECT MATTER

Independent claim 73 is directed to a ceramic support for supporting a catalyst component used, for example, for cleaning exhaust gas from an automobile's internal combustion engine. The ceramic support has a number of pores for directly supporting the catalyst component either within or at least partially within the pores of the ceramic support. Thus Appellants' invention provides a ceramic support for supporting a catalyst component without a coating of  $\gamma$ -alumina. The embodiment of claim 1 is shown in Figure 1 and described in the specification at, *inter alia*, p. 3, I. 34 – p. 4, I. 11.

Independent claim 80 is directed to a catalyst-ceramic body having a ceramic support with a cordierite honeycomb structure. The cordierite composition of the honeycomb structure replaces at least one of Si, Al, and Mg with a metal having a

catalyst activity. The embodiment of claim 80 is also shown in Figure 1 and described in the specification at, *inter alia*, p. 4, l. 12-16 and p. 23, l. 19 – p. 24, l. 1.

#### **GROUNDS OF REJECTION TO BE REVIEWED ON APPEAL**

Whether claims 73, 76-77 and 80-83 are anticipated under 35 U.S.C. § 102(b) and whether claims 74 and 75 are obvious over 35 U.S.C. § 103 by Guile et al. (U.S. Pat. No. 5,716,899).

Whether claims 73, 76-86, 97 and 102 are obvious under 35 U.S.C. § 103 over Ichii et al. (U.S. Pat. No. 5,607,885) in view of Beauseigneur et al. (U.S. Pat. No. 5,346,722).

Whether claims 94, 96 and 98-99 are obvious under 35 U.S.C. § 103 over Ichii et al. in view of Knapton et al. (U.S. Pat. No. 4,189,405).

Whether claim 95 is obvious under 35 U.S.C. § 103 over Ichii et al. in view of Abe et al. (U.S. Pat. No. 5,489,865).

#### WHETHER THE CLAIMS STAND OR FALL TOGETHER

With respect to the § 102 and § 103 rejections, claims 73 and 77 stand or fall together and claims 80-83 stand or fall together.

With respect to the other § 103 rejections, claims 73 and 76-77 stand or fall together. Claims 80-83, 85, 86 and 102 stand or fall together; and each one of claims 78, 84, 94, 95, 96, 97, 98 and 99 stands or falls alone.

### ARGUMENTS WITH RESPECT TO THE ISSUES PRESENTED FOR REVIEW

#### Rejection under 35 U.S.C. § 102

The Examiner's Final Office Action has improperly rejected claims 73, 76-77 and 80-83 as being anticipated under 35 U.S.C. § 102(b) by U.S. Patent No. 5,716,899 to Guile et al.

Under 35 U.S.C. § 102, a patent claim is invalid if it is anticipated by a single prior art reference. *Glaxo Inc. v. Novopharm Ltd.*, 52 F.3d 1043, 1047, 34 USPQ2d 1565, 1567 (Fed. Cir. 1995). To anticipate a patent claim, a prior art reference must disclose every limitation of the claimed invention, either explicitly or inherently. *In Re Schreiber*, 128 F.3d 1473, 1477, 44 USPQ2d 1429, 1431 (Fed. Cir. 1997).

Claim 73 clearly recites that a catalyst component is <u>directly supported</u> within or at least partially within the pores of the ceramic support. In rejecting this claim as being anticipated by Guile et al. the Examiner has improperly ignored this limitation, which the cited reference is not believed to teach or suggest.

Guile et al. discloses its catalytic material as a catalyst metal or catalyst metal oxide dispersed on an active material such as activated carbon or zeolite. Accordingly, Guile et al. does not disclose a catalyst directly supported on a ceramic body, as required by claim 73. Instead, Guile et al. discloses the use of an intermediate material such as activated carbon between the substrate and the catalyst metal. Accordingly, claim 73 and its dependent claims 76-78 are believed to patentably define over the cited reference.

At page 7 of the final Office Action the Examiner alleges that Appellants used open claim language, i.e., "comprising" to claim a <u>catalyst</u> "which allows for the

\* KOIKE et al Serial No. 09/546,227 November 22, 2004

possibility of more than just the claimed catalyst component to be supported on the substrate." Thus, the Examiner has misconstrued the claim which does <u>not</u> claim a catalyst, as alleged by the Examiner, but rather claims "[a] catalyst ceramic body comprising a ceramic support . . . and a catalyst component directly supported within, or at least partially within, the pores of said ceramic support." The latter limitation requires that the catalyst be directly supported within or partially within the pores of the ceramic support. Guile et al. does not meet this limitation because an active material, i.e., activated carbon or zeolite, is disposed between the catalyst and support.

In rejecting claim 80, the Examiner alleges that Guile et al. discloses "cordierite, Si, and AI (see column 3, lines 27-38 and 46)." Appellants respectfully disagree. Guile et al. describes some materials that are especially suited to the practice of its disclosed devices. More particularly, Guile et al. at column 3, lines 27-46 states:

The substrate materials are those that include as a predominant phase: ceramic, glass-ceramic, glass, cermet, metal, oxides, and combinations thereof. By combinations is meant physical or chemical combinations, e.g., mixtures, compounds, or composites. Some materials that are especially suited to the practice of the present invention, although it is to be understood that the invention is not limited to such, are those made of cordierite, mullitc, clay, talc, zircon, zirconia, spinel, alumina, silica, borides, lithium aluminosilicates, alumina silica, feldspar, titania, fused silica, nitrides, borides, carbides, e.g., silicon carbide, silicon nitride or mixtures of these. Some typical ceramic substrates are disclosed in U.S. Pat. Nos. 4,127,691 and 3,885,977. Those patents are herein incorporated by reference as filed. Especially suited substrate materials are cordierite, mullitc, and combinations thereof. Other types of bodies are porous metal bodies. Some preferred types of porous metal bodies, although it is to be understood that the invention is not limited to such, are bodies made of iron group metals such as, for example, Fe-Al or Fe-Cr-Al with optional additions for enhancement of various properties.

From the above, it should be clear that Guile et al. does <u>not</u> disclose "a honeycomb structure comprising at least as a main component a cordierite composition, wherein at least one of Si, Al and Mg elements constituting the cordierite composition being replaced by a metal having a catalyst activity" as required by present claim 80.

The Examiner has ignored the express claim language requiring that at least one element of Si Al and Mg being replaced by a metal having catalyst activity. At page 8 of the final Office Action the Examiner argues that the "replacing" limitation "appears to be a process step limitation rather than a product limitation" and therefore gives the limitation no patentable weight. To the contrary, the limitation provided a "catalyst-ceramic body" structure in which a catalytic material is included within the honeycomb structure and this structure is clearly not anticipated (or obvious over) the cited reference.

Accordingly, claim 80 and its respective dependent claims 81-86 and 102 also patentably define over the cited reference.

#### Rejections Under 35 U.S.C. § 103

Under 35 U.S.C. §103, a patent claim is invalid if the differences between its subject matter and the prior art are such that the subject matter as a whole would have been obvious at the time the invention was made to a person having ordinary skill in the art to which the subject matter pertains. *Litton Systems Inc. v. Honeywell Inc.*, 87 F.3d 1559, 1566, 39 USPQ2d 1321, 1325 (Fed. Cir. 1996).

The determination of obviousness is a question of law based on underlying factual inquiries. *Kegel Co. v. AMF Bowling Inc.*, 127 F.3d 1420, 1430, 44 USPQ2d 1123, 1130 (Fed. Cir. 1997). The ultimate determination as to obviousness is based on

four factual inquiries: the scope and content of the prior art, the differences between the claims and the prior art, the level of ordinary skill in the pertinent art, and secondary considerations, if any, of non-obviousness. *Id*.

#### A. Ichii et al. in view of Beauseigneur et al.

The Examiner has improperly rejected claims 73, 76-86, 97 and 102 as being unpatentable over Ichii et al. in view of Beauseigneur et al.

At page 3 of the final Office Action the Examiner states that Ichii et al. discloses, inter alia, "a honeycomb catalyst carrier without a coating (see column 1, lines 63-66)." However, the cited portion of Ichii et al. does <u>not</u> support the Examiner's assertion. Moreover, as Appellant explained in the Amendment filed on September 4, 2002, together with the Declaration of co-inventor Masakaza Tanaka (attached hereto as Appendix II), no mention of a coating in Ichii et al. does not mean that a coating is not necessary. In any event, the Examiner admits that Ichii et al. does not disclose a catalyst.

The Examiner alleges that Beauseigneur discloses pore diameter less than 5 microns (see column 3, lines 67-68) and catalyst metals (see column 6, line 29 and column 7, lines 65-68). However, Beauseigneur does not teach or suggest the features of independent claim 73 of the present application, namely, a catalyst directly supported within or partially within the pores of the ceramic support.

In rejecting the claims in the final Office Action, the Examiner states that Beauseigneur, *inter alia*, disclosed micro pores having a size of less than 5 µm. However, this disclosure is not relevant to Appellants' inventions as will be explained in

greater detail below. (With reference to the following patentability arguments, please see the sheet of figures attached hereto as Appendix III.)

Beauseigneur discloses a method for solving a problem associated with the provision of a washcoat layer for a catalyst. That is, Beauseigneur discloses that a coating layer having a large surface area such as a washcoat is necessary for a ceramic honeycomb. If the micro pores having a pore size less than 5 µm are effective to support a catalyst component, a washcoat layer is not necessary. Nevertheless, since a washcoat layer is formed in Beauseigneur, it clearly suggests that the micro pores of a ceramic honeycomb of Beauseigneur cannot support a catalyst component.

In Beauseigneur, the entry of a washcoat layer – comprising a catalyst component—into micro pores having a pore size of less than 5 μm is <u>prevented</u> by formation of a gel at the micro pores so as to provide a support having thermal shock resistance. Therefore, a catalyst component (catalyst particles), present in the washcoat layer is <u>not</u> even partially within the micro pores having a pore size less than 5 μm (see the upper figure of the attached sheet and the cited reference at Figure 3, column 2, lines 3-19, and column 3, line 64 through column 4, line 8).

In contrast, in Appellants' inventions, catalyst components are present within the fine pores of a support having a fine pore size of 100 nm or less (see the middle figure of the attached sheet and compare 5 µm with 100 nm or less). In Appellants' inventions, even if fine pores are less than 0.1 nm in size, a catalyst component can still be partially supported within the fine pores (see the lower figure of the attached sheet).

In rejecting claim 78, the Examiner alleges that Ichii discloses "microcracks on particles of 50 microns, which disappear (see Abstract and column 8, lines 52-55)."

However, as can be seen in the Abstract, Ichii discloses only particles (crystal diameter) of 50 microns or more, not microcracks of 50 microns or less. Similarly, the Examiner's reference to column 8, lines 52-55 (which refers to Table 1) merely describes particles having a crystal diameter of more than 30 microns but makes no mention of microcracks of 50 microns or less. Accordingly, claim 78 is believed to further patentably define over the cited art.

At pages 3-4 of the Office Action, the Examiner states "that it would have been obvious to one of ordinary skill in the art at the time the invention was made to use pore diameter, ceria, and metals of Beauseigneur in the honeycomb cordierite of <a href="Lichii">Lichii</a> because Beauseigneur discloses his ceria as particularly preferred (see column 6, lines 54-55), in a method of improving thermal shock resistance (title), in a honeycomb cordierite (see column 1, lines 5-24), to support catalyst metals for use as a catalyst (see column 7, lines 65-68)." However, it is respectfully submitted that Ichii does not disclose a honeycomb structure having at least one of oxygen vacancies and lattice defects in the cordierite crystal lattice on which a catalyst component is supported, as required in claim 84. The Examiner's reliance on Ichii at column 1, lines 63-66 for this teaching is clearly misplaced as this portion of Ichii makes no such mention of the above enumerated features of claim 84.

Nor can any relevant citations from the cited references be located for teaching the features of claims 80-83, 85, 86 and 102. As noted above, the Examiner combines Ichii with Beauseigneur, but in no way provides any reference as to which portions of the Ichii and Beauseigneur references are to be applied against these claims. With respect to independent claim 80 it appears that the Examiner has again improperly

ignored the limitation that the honeycomb structure has a cordierite composition in which a catalyst is provided in place of either Si, Al, or Mg. Since this limitation is not found in the cited art, claim 80 and its dependent claims 81-86 and 102 are believed to patentably define over the cited art.

In rejecting claim 97, the Examiner alleges that Ichii "discloses mixing and pouring both of which would cause vibration in a liquid." However, Appellants respectfully submit that Ichii only discloses <u>pouring a melt in water and does not disclose application of vibration during catalyst supporting</u> as required by claim 97. Accordingly, claim 97 is believed to further patentably define over the cited art.

The Examiner's rejection of claim 103 appears to be a typographical error since this claim has been allowed.

#### B. Ichii et al. in view of Knapton et al.

The Examiner has improperly rejected claims 94, 96 and 98-99 under 35 U.S.C. § 103(a) as being unpatentable over Ichii as applied to claim 73 above, and further in view of Knapton et al.

As noted above, the Examiner admits that Ichii alone does not invalidate claim 73. Since Knapton does not solve the deficiencies noted above with respect to Ichii et al., claims 94, 96 and 98-99 patentably define over the cited art. Moreover, Knapton discloses the use of CVD for forming an alumina layer, but does <u>not</u> disclose use of CVD for catalyst supporting. Accordingly, there is no reason to combine Ichii and Knapton, but even if the references were combined Appellants' inventions as recited in

claim 94 would not have resulted since the references do not teach or suggest using CVD (or PVD) to deposit a catalyst on the ceramic support.

In rejecting claim 96, the Examiner alleges that use of an organic solvent having a higher surface tension would have been obvious. However, claim 96 is directed to using an organic solvent having a lower surface tension than water. Accordingly, claim 96 is believed to further patentably define over the cited art.

With respect to claims 98 and 99 it is respectfully submitted that Knapton does not teach or suggest the process of production recited in these claims. More particularly, Knapton et al. does not teach or suggest first depositing a catalyst on the ceramic support and then following with a heat treatment. Accordingly, claims 98 and 99 are believed to further patentably define over the cited references.

#### C. Ichii et al. in view of Abe et al.

The Examiner has improperly rejected claim 95 under 35 U.S.C. § 103(a) as being unpatentable over Ichii as applied to claim 73 above, and further in view of Abe et al.

As noted above, the Examiner has admitted that Ichii alone does not invalidate claim 73. Moreover, Abe discloses using a supercritical state for drying a gel but does not disclose using a supercritical state for catalyst supporting. Therefore, the combination of a supercritical state disclosed in Abe with Ichii to constitute Applicants' invention, as recited in present claim 95, would not have been obvious. Hence claim 95 is believed to further patentably define over the cited art.

#### **CONCLUSION**

For all of the reasons set forth above, it is respectfully requested that this appeal be granted and that the rejection discussed above be reversed.

Respectfully submitted,

**NIXON & VANDERHYE P.C.** 

By:

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#### **APPENDIX I**

#### **APPENDIX OF CLAIMS ON APPEAL**

- 73. A catalyst ceramic body comprising a ceramic support having a number of pores and a catalyst component directly supported within, or at least partially within, the pores of said ceramic support.
- 74. The catalyst-ceramic body according to claim 73, wherein said catalyst-ceramic body comprises a catalyst component in an amount as the metal element of not less than 0.01% by weight, an average distance between particles of said catalyst component on the surface of said ceramic support being in a range of 0.1 to 1000 nm.
- 75. The catalyst-ceramic body according to claim 73, wherein an average distance between particles of said catalyst component is in a range of 0.1 to 100 nm.
- 76. The catalyst-ceramic body according to claim 73, wherein said catalyst component includes at least one component selected from the group consisting of metals having a catalyst activity and metal oxides having a catalyst activity.
- 77. The catalyst-ceramic body according to claim 76, wherein said metals having a catalyst activity are noble metals and said metal oxides having a catalyst activity are oxides containing at least one metal selected from the group consisting of V, Nb, Ta, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Zn, Ga, Sn, and Pb.

- 78. The catalyst-ceramic body according to claim 73, wherein said ceramic support has a multiple number of fine pores with a diameter or width of 0.1 to 100 nm on the surface of the ceramic support.
- 80. A catalyst-ceramic body comprising a ceramic support comprising a honeycomb structure and having a cordierite composition, wherein at least one of Si, Al and Mg elements constituting the cordierite composition being replaced by a metal having a catalyst activity.
- 81. The catalyst-ceramic body according to claim 80, wherein said catalyst-ceramic body contains a metal having a catalyst activity in an amount of not less than 0.01% by weight thereof.
- 82. The catalyst-ceramic body according to claim 80, wherein said catalyst-ceramic body contains a metal having a catalyst activity in an amount of not less than 0.01% by weight thereof and CeO<sub>2</sub> in an amount of not less than 0.01% by weight thereof.
- 83. The catalyst-ceramic body according to claim 80, wherein said metal having a catalyst activity includes at least one material selected from the group consisting of noble metals, V, NB, Ta, Cr, Mo, W, Mn, Fe, Co, Ni, Cu, Zn, Ga, Sn, and Pb.

- 84. The catalyst-ceramic body according to claim 80, wherein said honeycomb structure has at least one of oxygen vacancies and lattice defects in the cordierite crystal lattice, on which a catalyst component is supported.
- 85. The catalyst-ceramic body according to claim 80, wherein said honeycomb structure has a multiple number of fine cracks in at least one of the amorphous and the crystal phases thereof, on which a catalyst component is supported.
- 86. The catalyst-ceramic body according to claim 85, wherein said fine cracks have widths of not more than 100 nm.
  - 87. A process for producing a catalyst-body, comprising:

preparing cordierite materials comprising a Si source, an Al source and a Mg source as well as a binder, some of said Si, Al and Mg sources being replaced by a noble metal-containing compound,

forming said cordierite materials into a honeycomb shape,

heating said honeycomb shape to remove said binder, and

firing said honeycomb shape in a reduced pressure atmosphere at a pressure of not higher than 4000 Pa, a reducing atmosphere, an oxygen-containing atmosphere or an oxygen-free atmosphere to form a catalyst-ceramic body comprising a ceramic support of a honeycomb structure comprising a cordierite composition.

88. A process for producing a catalyst-ceramic body, comprising:

preparing cordierite materials comprising a Si source, an Al source and a Mg source as well as a binder, some of said Si, Al and Mg sources being replaced by a noble metal-containing compound and a Ce-containing compound,

forming said cordierite materials into a honeycomb shape,

heating said honeycomb shape to remove said binder, and

firing said honeycomb shape in a reduced pressure atmosphere at a

pressure of not higher than 4000 Pa, a reducing atmosphere, an oxygen-containing

atmosphere or an oxygen-free atmosphere to form a catalyst-ceramic body comprising

a ceramic support of a honeycomb structure comprising a cordierite composition.

- 89. The process according to claim 87, wherein said fired honeycomb structure is further heated to a predetermined temperature and then rapidly cooled from said predetermined temperature.
- 90. The process according to claim 87, wherein said fired honeycomb structure is further rapidly cooled to a predetermined temperature during cooling from a firing temperature.
- 91. The process according to claim 89, wherein a temperature different between said predetermined temperature and the temperature after said rapid cooling is not more than 900°C.

- 92. The process according to claim 87, wherein said fired honeycomb structure is further subjected to a shock wave.
- 93. The process according to claim 92, wherein said shock wave is provided by ultrasound or vibration.
- 94. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing at least one of a catalyst component and a precursor of a catalyst component on said ceramic support by one of a CVD and PVD method.
- 95. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing at least one of a catalyst component and a precursor of a catalyst component on said ceramic support by means of a super critical fluid.
- 96. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing at least one of a catalyst component and a precursor of a catalyst component on said ceramic support by means of a solvent having a surface tension smaller than water.
- 97. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing at least one of a catalyst component and a precursor of a catalyst component on said ceramic support by means of a solvent having a surface tension smaller than water while applying one of vibration and performing vacuum defoaming.

- 98. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing at least one of a catalyst component and a precursor of a catalyst component on said ceramic support by a heat treatment.
- 99. A process for producing the catalyst-ceramic body as set forth in claim 73, comprising depositing a catalyst component a plurality of times using the same or different catalyst compositions.
- 102. The catalyst-ceramic body according to claim 80, wherein said cordierite has a composition corresponding to a composition expressed by 2MgO /2•2Al<sub>2</sub>O<sub>3</sub>•5SiO<sub>3</sub>.
- 103. The process for producing a catalyst-ceramic body according to claim 89, wherein said cordierite has a composition corresponding to a composition expressed by 2MgO /2•2Al<sub>2</sub>O<sub>3</sub>•5SiO<sub>3</sub>.

#### **APPENDIX II**

#### **DECLARATION OF CO-INVENTOR MASAKAZA TANAKA**

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re application of:

T. KOIKE, ET AL.

oup art Unit:

Serial No.: 09/546,227

Examiner: JOHNSON, EDWARD M

1754

Filed: April 10, 2000

For: A CERAMIC SUPPORT CAPABLE OF SUPPORTING A CATALYST, A CATALYST-CERAMIC BODY AND PROCESSES FOR PRODUCING SAME

#### DECLARATION UNDER 37

Honorable Commissioner of Patents and Trademarks, Washington, D.C. 20231

#### Sir:

- I, Masakazu Tanaka, a citizen of Japan, residing at Urban-Life Sakae 501, Sakae-cho 4-62, Okazaki City, Aichi Pref. 444-0012, Japan, hereby declare the following.
- I am a co-inventor of the above-identified patent application.
- I obtained a masters degree from the Department of Chemical Engineering, Graduate School of Engineering, Kyushu University, where I studied fluid dynamics of mixture and/or I entered Nippon-Denso (now Denso Corporation) in agitation.

1987, and was appointed to the Research & Development
Department where I was engaged in the development of an
electric control suspension system for automotive use from
1987 to 1990, development of an electrically controlled tire
air pressure system for automotive use from 1990 to 1993, and
development of an exhaust gas purification system for
automotive use from 1993 to 1998. I was then appointed to
the Ceramic Engineering Department where I was engaged in the
design and development of a ceramic honeycomb substrate for a
catalyst converter.

3. As a person skilled in the art of automotive and ceramic honeycomb substrates for catalyst converters, I know that a catalyst is never supported on known ceramic supports when the catalyst is used in practice since the ceramic supports do not have a sufficiently high surface area. A high surface area material such as gamma-alumina is coated on a ceramic support such as a honeycomb structure and a catalyst is supported on the coated high surface area material. Without such a coating, a ceramic support cannot effectively support a sufficient amount of a catalyst directly thereon and such a catalyst-directly-supported ceramic support cannot be used as an affective catalyst or an affective catalyst-ceramic body.

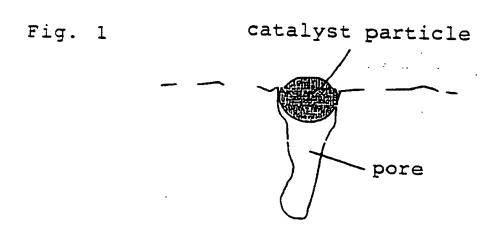
I also know that it is a common practice that in

much literature, coating is not mentioned even though a catalyst must have been supported on a ceramic support, and that a person skilled in the art therefore understands that even if the coating was not mentioned in literature, a coating was actually formed on a ceramic support. I can almost surely guarantee the above knowledge as a person who has had been concerned with and worked with the technology of catalyst supporting. At least it is certain that failure to mention a coating does not necessarily mean that a coating was not formed, and even though the coating is not mentioned in Ichii'885, a person skilled in the art would not consider that no coating was formed in Ichii'885.

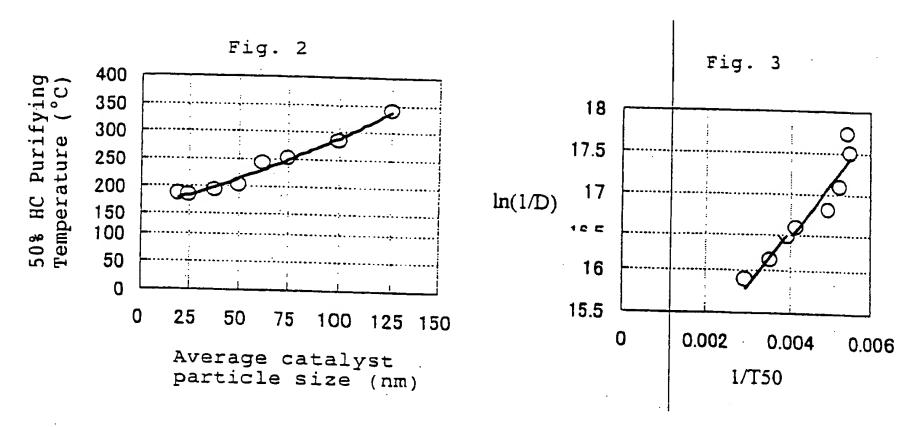
- 4. Next, I will show that a ceramic body having fine pores with a size of 0.1 to 100 nm can effectively support a catalyst but a ceramic body having pores with a size of more than 248 cannot effectively support a catalyst.
- 5. As shown in the following Fig. 1, it can be considered that the size of fine pores of a ceramic body corresponds to the size of a catalyst particle which can be supported by the fine pores. If the size of the catalyst particle is larger than the size of the fine pore, the catalyst particle cannot be fixed or supported by the fine pore. If the size of the catalyst particle is smaller than the size of the fine pore, the catalyst particle enters into

- 3 -

a deep portion of the fine pore and cannot effectively work as a catalyst since the catalyst particle does not contact a material to be catalyzed, for example, an exhaust gas to be purified.



6. Using a cylindrical catalyst sample comprising activated alumina and catalyst particles and having a diameter of 15 mm and a length of 10 mm and a hydrocarbon sample gas, the temperature (T50) at which 50% of the hydrocarbon sample gas was purified was measured, while the particle size of the catalyst particle was varied. The mean particle size (D) of the catalyst particles was measured by the CO pulse adsorption method. The obtained result is shown as T50v. D in the following Fig. 2.



It is known that T50 and D have the following relationship:

$$ln(1/D) = A/(T50) + B$$
 (1)

Here Fig. 2 is reduced to Fig. 3 which shows the relationship between  $\ln(1/D)$  and (1/T50). Fig. 3 shows that the formula (1) is satisfied. When the constants A and B in the formula (1) are obtained from Fig. 3, A = 645.45 and B = 13.891. That is, the formula (1) can be expressed as:

$$ln(1/D) = 645.45/(T50) + 13.891$$
 (1)

When no catalyst was used, T50 was measured to be 489°C.

This value of T50 =  $489^{\circ}$ C is inserted into the formula (1), the mean catalyst particle diameter (D) becomes 248 nm.

6. Therefore, it can be said that catalyst particles having a mean particle diameter (D) of larger than 248 nm exhibit substantially no catalyst performance.

In contrast, Fig. 3 shows that catalyst particles having a mean particle diameter (D) of 100 nm exhibit a sufficient catalyst performance as T50 is about 300°C.

7. As described above, since the fine pore size can be considered to be the diameter of supportable catalyst particles, fine pores having a mean particle diameter (D) of 100 nm can support catalyst particles having a sufficient catalyst performance, but pores having a mean particle diameter (D) of larger than 489 nm can hardly support catalyst particles having a sufficient catalyst performance.

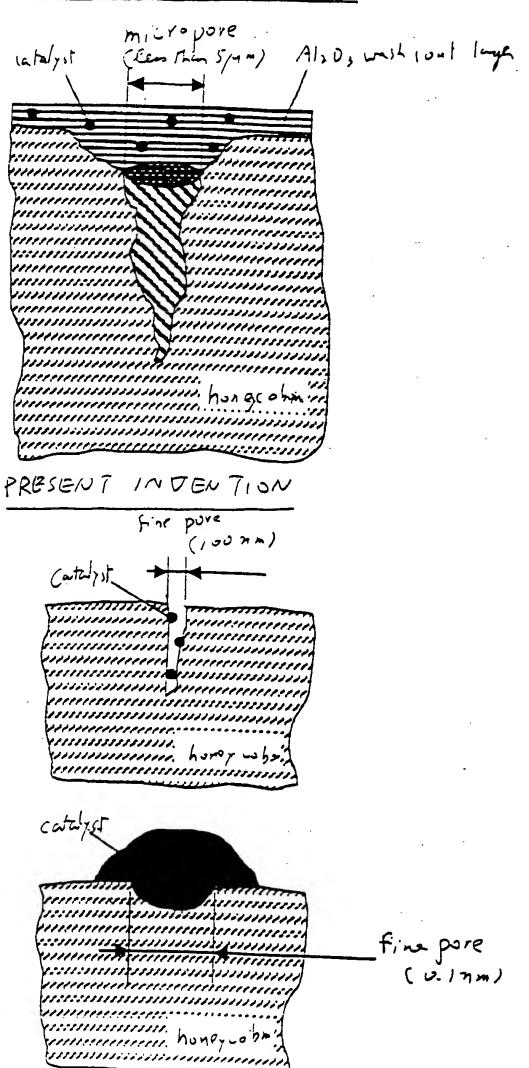
I, the undersigned declarant, declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true, and; further, that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under section 1001, of Title 18, of the United States Code, and that such willful false statements may jeopardize the validity of the application or any patent issuing thereon.

Signed this 9th day of August , 2002

Masakazu Tanaka

# APPENDIX III SHEET OF FIGURES

### US5346722 (Beaunseigneur)



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